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Will black carbon mitigation dampen aerosol indirect forcing?

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[1] If mitigation of black carbon (BC) particulate matter is accompanied by a decrease in particle number emissions, and thereby by a decrease in global cloud condensation nuclei (CCN) concentrations, a decrease in global cloud radiative forcing (a reverse "cloud albedo effect") results. We consider two present-day mitigation scenarios: 50% reduction of primary black carbon/organic carbon (BC/OC) mass and number emissions from fossil fuel combustion (termed HF), and 50% reduction of primary BC/OC mass and number emissions from all primary carbonaceous sources (fossil fuel, domestic biofuel, and biomass burning) (termed HC). Radiative forcing effects of these scenarios are assessed through present-day equilibrium climate simulations. Global average top-of-the-atmosphere changes in radiative forcing for the two scenarios, relative to present day conditions, are $+0.13 \pm 0.33$ W m^{-2} (HF) and $+ 0.31 \pm 0.33$ W m^{-2} (HC). **Citation:** Chen, W.-T., Y. H. Lee, P. J. Adams, A. Nenes, and J. H. Seinfeld (2010), Will black carbon mitigation dampen aerosol indirect forcing?, *Geophys. Res. Lett.*, *37*, L09801, doi:10.1029/2010GL042886.

1. Introduction

[2] Black carbon (BC) both absorbs and scatters radiation, resulting in heating of the atmosphere and reduction of solar radiation reaching the Earth's surface. Ambient measurements indicate that freshly emitted BC eventually becomes co-mixed with other compounds, such as sulfate, which are invariably less absorbing, but more hygroscopic, than BC. Whereas particles containing a mixture of BC and non-absorbing, but hygroscopic, components absorb proportionately more solar radiation than BC particles alone, the lifetime of such particles against wet deposition decreases relative to that of pure BC, leading to a complex interplay between enhanced radiative absorption but shorter lifetime [*Stier et al.*, 2007]. Accounting for the relative increase of BC since pre-industrial conditions allows reconciliation between satellite-derived and modeled estimates of direct aerosol forcing [*Myhre*, 2009]. Mitigation of BC leads to a reduction in (positive) top-of-the-atmosphere (TOA) direct radiative forcing, and therefore has been suggested as a strategy complementary to reduction of greenhouse gases [*Jacobson*, 2002; *Bond*, 2007].

[3] Any perturbation that affects the global aerosol number concentration has the potential to alter Cloud Condensation Nuclei (CCN) concentrations and cloud properties. The present study is based on two key assumptions: (1) a decrease in BC mass emissions would be accompanied by a decrease in primary particulate number emissions, which would lead to a lower global aerosol number concentration; and (2) by virtue of internal mixing with hydrophilic aerosol components, BC is assumed to contribute to the CCN population. Depending on the fraction of BC primary particles that eventually become CCN (a function of their growth and loss rates [*Pierce and Adams*, 2007]), BC mitigation would affect global CCN concentrations, leading to a change in global cloud radiative forcing from warm clouds. If such a perturbation were to result in a reduction in TOA cloud radiative forcing, the amount of that reduction would oppose the amount by which the TOA direct BC radiative forcing is also reduced.

[4] The "aerosol indirect effect" (AIE) generally relates to mechanisms by which aerosols affect cloud radiative forcing via the availability of CCN, through their size distribution, composition, and mixing state. The AIE has been divided according to: (1) the cloud albedo effect; and (2) the cloud lifetime effect. When both cloud albedo and lifetime effects are considered, the magnitude of the cloud lifetime effect has been found to range from 0.2 to 1.4 of that of the cloud albedo effect [*Lohmann and Feichter*, 2005]. Although the separation into these two effects has served as a useful way to describe aerosol-cloud interactions, it is widely appreciated that such a separation is an oversimplification [*Feingold and Siebert*, 2009; *Stevens and Feingold*, 2009]. For the purpose of the present study, we follow an approach to estimating aerosol indirect forcing similar to that of IPCC [*Forster et al.*, 2007].

2. Effect of BC Emission Reductions on Aerosol Indirect Forcing

[5] The present-day direct and indirect aerosol forcings associated with the prescribed reduction in BC emissions have been evaluated by employing the Goddard Institute for Space Studies (GISS) Global Climate Middle Atmosphere Model III (referred to as GISS III hereafter), following *Chen et al.* [2010] (see footnotes in Table 1). Effects from the changes in both cloud albedo and lifetime are considered, while indirect effects on convective clouds and mixed-phase and ice clouds are not included in the present study.

[6] The atmospheric aerosol number concentration is established, in principle, through a combination of primary particle emissions, in-situ nucleation, coagulation, and removal processes [*Adams and Seinfeld*, 2002; *Pierce and*

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Table 1. Scenarios and Results

	Aerosol					Cloud	
Scenario	Emission[a]	Source[b]	Burden[c]	Lifetime (days)	ADE Forcing (W m^{-2})[d]	N_c in Surface (cm^{-3})[c]	AIE Forcing (W m^{-2})[e]
BASE							
BC	7.7 Tg yr^{-1}	7.7 Tg yr^{-1}	0.17 Tg	7.8			
OC	60.6 Tg yr^{-1}	79.7 Tg yr^{-1}	1.1 Tg	5.2	—	196	—
N_a	2.26×10^4 cm^{-3} yr^{-1}	2.79×10^4 cm^{-3} yr^{-1}	2.83×10^2 cm^{-3}	5.3			
HF							
ΔBC	−19.7%	−19.7%	−21.4%	−0.1	−0.07 ± 0.0017 (int.)	187	
ΔOC	−3.6%	−2.7%	−3.0%	0	−0.03 ± 0.0002 (ext.)	(−5.9%)	+0.13 ± 0.33
ΔN_a	−22.5%	−17.3%	−4.2%	+0.5			
HC							
ΔBC	−50.0%	−50.0%	−49.7%	+0.1	−0.12 ± 0.0017 (int.)	179	
ΔOC	−50.0%	−38.0%	−35.8%	+0.2	−0.04 ± 0.0008 (ext.)	(−9.8%)	+0.31 ± 0.33
ΔN_a	−26.2%	−20.0%	−4.9%	+0.7			

[a]The change in primary particles listed is for *all* primary particles. Primary sulfate (from in-plume nucleation processes), sea salt, and mineral dust are unchanged in each of the scenarios. Fossil fuel emitted particles are ~ 45% by number of total primary particle emissions, so that a 50% reduction in that amount leads to a ~ 23% reduction by number in total particle emissions.

[b]The difference between the "Source" and "Emission" columns represents secondary production: SOA in the case of OC and nucleation in the case of particle number. The nucleation source is defined as J_{10}, the number flux of nucleated particles that grow past 10 nm diameter.

[c]In the BASE simulation, emissions from fossil fuel and biofuel sources are based on the emissions inventory of *Bond et al.* [2004], and biomass burning emissions are from the GFED inventory. Monthly-averaged concentrations of ammonium sulfate, OC, BC, and sea salt aerosols are derived from the TwO-Moment Aerosol Sectional microphysics model (TOMAS) [*Pierce and Adams*, 2009]. The aerosol activation parameterization of *Nenes and Seinfeld* [2003] and *Fountoukis and Nenes* [2005] is used to calculate corresponding monthly average N_c fields from a detailed global aerosol model [*Lee et al.*, 2009]. Updraft velocities representative of stratiform clouds (0.15 m s^{-1} over ocean and 0.3 m s^{-1} over land) are assumed.

[d]Direct aerosol forcing is determined by the instantaneous change in annual mean TOA net (shortwave plus longwave) radiative fluxes using GISS-III [*Chen et al.*, 2010]. Each simulation is integrated for two years with prescribed monthly mean sea-surface temperature (SST) from HadISST1 observed climatology for 1992 to 2000 [*Rayner et al.*, 2003]. Radiative forcings for internal (int.) and external (ext.) aerosol mixtures are derived separately. A standard gamma size distribution is assumed for the aerosol with a surface area-weighted dry radius (0.1 μm for pure BC in external mixture; 0.3 μm for all other species and the internal mixture) and variance (= 0.2). The wavelength-dependent refractive indices of dry sulfate, nitrate (assumed the same as that of sulfate), and sea salt are taken from *Toon et al.* [1976], with those for organic carbon, BC, and water from *d'Almeida et al.* [1991]. Extinction efficiency, single-scattering albedo, and asymmetry parameter are then supplied to the radiation scheme of the GISS III [*Schmidt et al.*, 2006; *Rind et al.*, 2007] to calculate aerosol optical depth and radiation fluxes. For an internal mixture, the density of the aqueous aerosol mixture is computed as the mass-averaged density of water and dry aerosols, and the composite aerosol radiative properties are derived based on a homogeneous volume-weighted mixing rule. For external mixing, the Mie calculation is applied for individual species of dry aerosols and aerosol water.

[e]To derive the AIE forcing, modifications have been made by *Chen et al.* [2010] to the formulations of optical depth and autoconversion rates in liquid-phase stratiform clouds in GISS III to introduce explicit dependence on N_c fields. Following the common measure of AIE forcing [*Forster et al.*, 2007], the cloud properties and hydrological cycle are allowed to respond accordingly, and the AIE forcing is determined by the change in annual mean TOA net (shortwave plus longwave) cloud forcing (all-sky minus clear-sky radiative fluxes) between each set of simulations. Each simulation is integrated for 20 years with prescribed SST and specific levels of offline, monthly-averaged N_c values from the TOMAS model. With the first five years as a spin-up period, the average AIE forcing over the last 15 years is reported.

Adams, 2009]. The effect of BC mitigation on global CCN concentrations is assessed using the GISS-TOMAS model [*Adams and Seinfeld*, 2002]. Primary particles considered include sea salt, mineral dust, "primary" sulfate produced in power plant plumes, and primary BC and organic carbon (OC). Sea salt includes a portion of ultrafine particles [*Pierce and Adams*, 2006]. Atmospheric new particle formation is represented by binary H_2SO_4-H_2O nucleation, accounted for using the model of *Vehkamäki et al.* [2002]. Particle growth occurs by coagulation and condensation of sulfate and secondary organic aerosol (SOA). Nitrate formation is not included in the present calculation, as it is likely to exert a small effect under present-day conditions.

[7] According to Köhler theory, the CCN activity of a chemically heterogeneous particle depends primarily on the moles of solute it contains. Since the majority of the emitted particles are small, ultrafine particles and initially hydrophobic, the amount of solute they contain will be dominated by sulfuric acid and organics that condense on them during atmospheric processing. For example, even modest growth of an initially hydrophobic diesel particle from 80 nm to 100 nm implies that its volume has approximately doubled. The solute added by the condensed sulfate and organics will completely dominate the ~1% co-emitted sulfate of the initial soot particle. Put another way, Köhler theory predicts that a particle with 1% sulfate would need to have a diameter of ~350 nm in order to activate at supersaturations of ~0.2%. A negligible fraction of the emissions of interest occur at that size, and sulfate added to soot particles by atmospheric processing (condensation) is critical to their CCN activity.

[8] For a soot particle to function as a CCN: (1) A soot particle must become internally mixed with other aerosol components (e.g., sulfate and organics), and this is assumed to occur with a 1.5 day aging timescale. Once internally mixed with other, soluble aerosol components, the particle may or may not be a CCN depending on its size and the overall chemical composition of that size bin in that model grid cell; and (2) Once "internally mixed", the typical soot particle will still be quite small (e.g., 20–60 nm) and therefore will not activate regardless of its composition. The particles then have to grow by condensation to the sizes (~100 nm diameter) where they will activate and function as CCN [*Adams and Seinfeld*, 2002; *Pierce and Adams*, 2009]. Under essentially all reasonable atmospheric conditions, the second step is the limiting one for a soot particle to become a CCN. In the BASE simulation, ten bins span the size range from 10 nm to 1 μm dry diameter with an additional two size bins in the coarse mode. Mineral dust is included according to the treatment of *Lee et al.* [2009], and biomass

Table 2. Global Sources of Aerosol Number Concentrations[a]

Process	Rate (particles cm^{-3} yr^{-1})
"Primary" (plume nucleation) sulfate[b]	1.02×10^4
Sea-salt emissions	5.4×10^2
EC/OC from fossil fuel[b]	1.02×10^4
EC/OC from biofuel/biomass	1.66×10^3
Mineral dust emissions	2.08×10^1
Nucleation (J_{10})	5.24×10^3

[a]Particles larger than 10 nm.

[b]That the magnitudes of these two rates are the same is coincidental. The sulfate mass emission rate is ~ 2.1 Tg sulfate yr^{-1}, and the EC/OC fuel emission rate is ~ 7.2 Tg yr^{-1}, a ratio of 0.29. The number emission rate per kg of primary sulfate is 3.57 times that per kg of EC/OC fossil fuel. Then, 0.29 × 3.27 ~ 1.

burning emissions are based on the GFED inventory [*van der Werf et al.*, 2004].

[9] BC particles can act as ice nuclei [*Phillips et al.*, 2008], impacting the microphysical state of clouds at temperatures below the freezing point of liquid water. In mixed-phase clouds, decreases in BC ice nuclei concentrations may reduce the glaciation frequency of supercooled clouds, which can decrease precipitation and enhance shortwave cloud albedo and longwave emissivity [*Lohmann and Feichter*, 2005]. BC reduction in cold cirrus clouds may enhance crystal number concentrations, especially if both heterogeneous and homogeneous freezing are actively competing for water vapor [*Barahona and Nenes*, 2009ab]. Ice crystal concentration enhancements will increase the longwave emissivity of cirrus [*Karcher et al.*, 2007]. These ice nuclei effects on climate are not considered in this study.

[10] The scenarios considered consist of three cases (Table 1):

[11] BASE: present-day conditions; primary emissions from fossil fuel combustion are assumed to follow a log-normal size distribution with number median diameter = 25 nm and geometric standard deviation = 2.0; biofuel and biomass burning emissions have number median diameter = 100 nm and geometric standard deviation = 2.0. (To represent coagulational scavenging of fresh emissions on subgrid scales, the fresh primary size distributions undergo 10 h of subgrid processing before being added to model grid cells [*Pierce et al.*, 2009]).

[12] HF: "half fossil fuel"; primary BC/OC mass and number emissions from fossil fuel combustion are each reduced by 50%.

[13] HC: "half carbonaceous"; primary BC/OC mass and number emissions from all primary carbonaceous sources (fossil fuel, domestic biofuel, and biomass burning) are reduced by 50%; this scenario represents deeper reductions than HF; note that emissions reductions in this scenario occur mostly at particle sizes larger than those of HF.

[14] Table 1 gives the annual mean aerosol budgets for the BASE, HF, and HC scenarios. In the HF scenario, BC mass emissions are reduced by ~20%. Since the fossil fuel emissions inventory is predominantly BC, with only small amounts of OC, the total emissions of OC in HF decrease by only about 4%. Global OC emissions are dominated by biomass burning and domestic biofuel combustion. The change in particle number emitted in scenario HF is also relatively high (~23%) because the fossil fuel particles are relatively small (~25 nm). In contrast, the HC scenario has (by definition) a 50% reduction in primary BC and OC mass emissions. Although this represents much deeper cuts in BC/OC mass than the HF scenario, primary number emissions are reduced only modestly further (26% in HC vs. 23% in HF) because the biomass burning emissions involve larger particles (~100 nm). The fact that only a 26% reduction in total primary particle number occurs, even in the HC scenario, is a reflection of the fact that the global aerosol number concentration comprises many non-carbonaceous particles. Mineral dust is a small contribution. Ultrafine sea salt is a somewhat larger contribution, and primary sulfate is a large contributor since its assumed size distribution peaks ~20 nm.

[15] The sources of global aerosol number concentration (for particles larger than 10 nm) are given in Table 2. Note that in Table 2, "nucleation" refers to the number of particles that grow to 10 nm diameter and are, therefore, tracked explicitly in the model. The formation rate of critical clusters (~1 nm diameter) is much faster. Biomass and biofuel combustion are ~6% of the total source of aerosol number. Natural particles (sea-salt plus mineral dust) are ~2% of the total source of aerosol number, a value that includes a substantial contribution from ultrafine sea-salt emissions. The fraction of biomass burning that is natural is uncertain, but generally thought to be small (~10%), so the ~2% does not include any contribution from biomass burning.

[16] Generally, only a small fraction of primary combustion particles is smaller than 10 nm. Typically, atmospheric particles smaller than 10 nm result from homogeneous nucleation. Particles smaller than 10 nm, however, are extremely short-lived in the atmosphere (see, for example, *Zhang et al.* [2004]). For purposes of CCN calculations, neglect of particles smaller than 10 nm has only a minor impact. The probability of these particles growing to be CCN is very low since they undergo efficient coagulational scavenging in the atmosphere [*Pierce and Adams*, 2007] and even much larger changes in the nucleation rate of particles smaller than 10 nm has a relatively minor impact on global CCN [*Pierce and Adams*, 2009]. Finally, for the purposes of emissions book-keeping in Table 1, the 10 nm cutoff is somewhat arbitrary. It represents, however, a reasonable balance between including most primary particles and excluding those that are too short-lived to influence CCN.

[17] Particle emissions from fossil fuel sources are sometimes reported as bimodal, where the smallest mode contains organic/sulfate particles and the second mode contains black carbon/organic/sulfate particles. Here we have assumed a single mode. We performed sensitivity simulations to investigate the effect of this assumption. Comparing the two cases, the resulting change in global cloud drop number concentration between unimodal to bimodal cases is ~2%.

[18] Global average present-day AIE forcing at TOA for the two scenarios relative to the BASE case are: + 0.13 ± 0.33 W m^{-2} (HF) and + 0.31 ± 0.33 W m^{-2} (HC). The magnitude of these (positive) forcings can be compared to the best guess IPCC [*Forster et al.*, 2007] value of pre-industrial minus present-day AIE forcing from the cloud albedo effect of −0.7 W m^{-2}. The standard deviations associated with the predicted values for the two scenarios are a result of the inter-annual variability of cloud and precipitation. Reduction of BC leads also to decreased heating of

the atmosphere via shortwave absorption. The TOA aerosol direct forcing (assuming an internal aerosol mixture) associated with the two scenarios is: −0.07 W m^{-2} (HF) and −0.12 W m^{-2} (HC). These direct aerosol forcings due to BC control can be compared with the IPCC estimated present-day minus pre-industrial direct radiative forcings for fossil fuel and total BC/OC of +0.15 and +0.18 W m^{-2}, respectively. Noting that direct forcing is calculated by diagnosing the change in TOA radiative fluxes in the absence of feedbacks, whereas indirect forcing is calculated from the perturbation in cloud forcing (all-sky minus clear-sky fluxes) allowing for feedbacks in cloud water and precipitation, we can estimate the net climatic effect of the two scenarios as: +0.06 W m^{-2} (HF) and +0.19 W m^{-2} (HC).

3. Emissions Scenarios

[19] In the HF and HC emissions scenarios examined here both the mass and number of emitted carbonaceous particles are reduced from their respective sources by 50%. These scenarios also assume that the size distribution of emitted particles remains invariant. In reality, the change in the size and number of emitted particles will depend on the precise controls adopted [*Kittelson*, 1998]. Recent measurements on a suite of particulate matter control technologies for heavy-duty diesel vehicles found that number emission factors (particles km^{-1}) could increase approximately one order of magnitude or decrease by ~3 orders of magnitude depending on the control technology in question [*Biswas et al.*, 2008].

[20] The relatively simple scenarios considered here can be considered a reasonable starting point for analysis. First, some control strategies (e.g., replacement of domestic biofuel burning by a much cleaner alternative) lead to proportional reductions in both mass and number emissions. Second, concerns over potential health impacts of ultrafine particles may lead to a preference for control technologies that reduce both mass and number. Third, measurements made in a tunnel near San Francisco in 1997 and 2006 indicate that number emission factors for both light-duty vehicles and diesel trucks have decreased over that time period by a proportion similar to, but somewhat less than, the mass emission factor decrease [*Ban-Weiss et al.*, 2010], although these data do not yet reflect widespread deployment of diesel particulate filters. Nevertheless, one cannot rule out an increase in number emissions if BC controls are implemented; this would likely lead to an enhancement of the aerosol indirect effect, precisely the opposite of the result obtained here.

4. Conclusion

[21] Black carbon emission control is recognized to be an effective strategy for mitigation of both global warming and air pollution. If BC mass emission reduction is reflected in an alteration of both the global CCN concentration and cloud droplet number concentration, such a reduction can lead potentially to a change in indirect aerosol radiative forcing. We have evaluated the change in aerosol indirect forcing under present-day conditions for two scenarios of BC control: (1) 50% reduction in fossil fuel emissions of BC/OC; and (2) 50% reduction in BC/OC emissions from all primary carbonaceous aerosol sources. The estimated net changes in TOA cloud radiative forcing corresponding to the two scenarios are +0.13 and +0.31 W m^{-2}, respectively, that is, a reduction in global cloud radiative forcing. Because of the importance of both BC control and of understanding aerosol indirect effects, additional studies of the interplay between these two issues are warranted.

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